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# EXPERIMENTAL RESULTS ON HIGH-ENERGY PARTICLE PRODUCTION OF $Na^{24}$ IN CONCRETE

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## I. INTRODUCTION

After either the booster of main-synchrotron accelerator is shut down, the main sources of residual radiation in the enclosures will be the steel in the magnets and the concrete walls of the enclosure. The activity induced in the accelerators will have to be accepted as an undesired but unavoidable part of accelerator operation. The activity created in the concrete consists almost exclusively of $Na^{24}$, which has a 15 hour half-life, and which yields 2.75 and 1.37 MeV gamma rays. The amount of $Na^{24}$ created in the concrete by thermal neutron capture by $Na^{23}$ and by spallation depends strongly on the chemical composition of the concrete. By controlling the chemical composition of the concrete it is possible to reduce the exposure rate in the machine enclosure from the residual activity in the concrete to a small fraction of the corresponding exposure rate from the accelerator itself.

Rough calculations of the exposure rate contribution of each of

several elements in concrete were made and reported in 1968.[1] The results of an experiment carried out in early 1969 at ANL to check these rough calculations and provide a firmer basis for evaluating different concrete mixes are reported here. The results of a more careful calculation[2] and another experimental result[3] have appeared recently.

## II. EXPERIMENTAL PROCEDURE

To simulate the effect of proton losses in a synchrotron we used the flux of particles coming from the ZGS during its normal operation. Samples were positioned in the accelerator enclosure downstream from the extraction target, where they were exposed to secondaries (mainly neutrons) emerging from the target at about 45° from the beam direction. The only material between the target and the samples was the 1-in. thick steel wall of the target box. Measurements of the neutron flux at the sample location using moderated gold foil detectors indicated that the average flux was about $4\times10^{6}$ n/cm$^{2}$-sec.

A concrete block, 3 ft × 3 ft × 1 ft deep was used to simulate a section of enclosure wall. Samples were placed at various depths in a 4-in. diameter cylindrical hole in the block for irradiation. The axis of the hole was pointed at the extraction target. A second, solid concrete block was placed behind the first to prevent scattered neutrons from entering the sample cylinder from the back end.

The following types of samples were irradiated:

1. Concrete disks of presumably known chemical composition

2. Reagent-grade chemicals containing Al, Mg, Ca, Si, and Na, the principal constitutents of concrete with $A \geq 23$

3. "Mock concrete" composed of a mixture of suitable proportions of the above chemicals

4. Sodium (NaCl) samples placed in between concrete disks.

The concrete disks, in addition to being studied as samples, also were used as fillers in between other samples. The powdered chemical samples were contained in either petri dishes or small medicine vials. The 4-in. diameter petri dishes filled the entire cross section of the sample hole, but different samples had to be placed at different depths. On the other hand, many small medicine vials could be placed at the same depth, but they did not completely fill the cross section of the hole.

The following activation reactions were used to monitor the neutron flux during the different runs:

$Al^{27}$ (n, $\alpha$) $Na^{24}$

Moderated gold foil: $Au^{197}$ (n, $\gamma$) $Au^{198}$

$Cu^{63}$ (n, $\gamma$) $Cu^{64}$ (bare and Cadmium-covered).

All detectors and samples were counted using a 4-in. × 4-in. NaI detector belonging to the ANL Health Physics Group.

The contributions of the Al, Mg, Ca, and Si to the concrete activation were determined by activating two or three samples of each element at different depths in the concrete block. If the fast neutron

flux responsible for producing $Na^{24}$ by spallation is attenuated exponentially with depth in the block, then the $Na^{24}$ specific activity will also fall off exponentially with depth:

$$A(x) = A_0 \exp(-x/x_0). \quad (1)$$

The parameters $A_0$ and $x_0$ were determined for each target element from the observed specific activity distributions.

The gamma exposure rate next to such a wall (or anywhere inside a uniformly irradiated cavity) can then be written as[4,5]

$$DR = A_0 d(E) \int_0^W B_r(\mu x, E) \exp(-\mu x) \exp(-x/x_0) dx, \quad (2)$$

where W is the wall thickness ($g/cm^2$), E is the gamma energy, d(E) is the dose per photon, and $\mu(E)$ and $B_r(\mu x, E)$ are gamma-ray attenuation constants and dose buildup factors. Equation (2) was numerically integrated and the result divided by the neutron flux to give the "activity coefficients" (exposure rate per incident neutron) in Table I.

The $Na^{23}$ (n, $\gamma$) $Na^{24}$ reaction proceeds by the capture of thermal rather than fast neutrons. Since the incident spectrum is hard, one would not expect the reaction rate to fall off exponentially with depth in the wall. Rather, the reaction rate will first increase with depth as the incident neutrons are thermalized, and will then decrease.

This is in fact what was observed when small Na samples were placed between concrete disk moderators and irradiated. The observed

specific activity was numerically integrated to obtain the resultant exposure rate outside the wall, as was done with Eq. (2).

In all cases the incident neutron flux was measured using moderated gold foil detectors, with the other detectors used for consistency checks. One of the problems encountered was that the incident spectrum changed whenever the ZGS operating or targeting conditions changed. This forced us to discard some of our early data.

The samples of "mock concrete" were used as consistency checks to see if the activity of the mexture was equal to the sum of the activities of the various components. The average discrepancy was 5%.

It was not possible to obtain agreement between the measured activities of the concrete samples and the predictions made on the basis of its chemical analysis (supplied by DUSAF), and the activation measurements made on the separate elements. This is not too surprising, as it is very difficult to obtain an accurate analysis of the sodium content of concrete.[6] In addition, the size of the aggregate used was such that the 4-in. diameter disks were not representative of the average composition of the concrete.

## III. RESULTS AND DISCUSSION

The results are most usefully presented in terms of an "activity coefficient," $a_i$, for each element. The exposure rate from $Na^{24}$ in concrete after an infinite irradiation time ($T_{on} >> 15$ hrs) and zero decay time is then

$$DR = \Phi \sum_i a_i f_i, \qquad (3)$$

where $\Phi$ is the incident neutron flux. The summation extends over the elements Al, Mg, Si, Ca, Na, and $f_i$ is the fraction by weight of each of the above elements in the concrete in question.

Table I.

| Element | $a_i$, (rad/hr)/(n/cm$^2$sec) | $f_i$ (Booster) |
|---|---|---|
| Al | $1.9\times10^{-8}$ | 0.0063 |
| Mg | $1.9\times10^{-8}$ | 0.099 |
| Ca | $0.016\times10^{-8}$ | 0.23 |
| Si | $0.33\times10^{-8}$ | 0.023 |
| Na | $60\times10^{-8}$ | 0.002 |

The uncertainties in the $a_i$ are typically 20%. The $f_i$ given above are average values for the Booster concrete.[7]

Extrapolation of the calculations of Armstrong and Alsmiller[8] from 3 to 8 GeV shows that when a proton is lost in a 40 g/cm$^2$ magnet, a total of 9.4 neutrons emerge from the magnet. The loss of one proton/cm-sec thus results in an average neutron flux at the enclosure walls of $\phi' = 0.13$ n/cm$^2$-sec. The anticipated average losses during acceleration will be[9]

$$dI/d\ell = 5.4\times10^5 \text{ p/cm-sec},$$

yielding a neutron flux at the walls of

$$\Phi = \phi \times \frac{dI}{d\ell} = 1.3\times10^5 \text{ n/cm}^2\text{sec}.$$

The average gamma exposure rate from $Na^{24}$ in the concrete enclosure walls will then be

$$DR = \Phi \sum_i a_i f_i = 0.43 \text{ mrad/hr.}$$

The calculation of Armstrong and Barish[2] gives

$$DR = 0.61 \text{ mrad/hr}$$

for the present concrete composition. Differences between the present activity coefficients and those of other measurements[3] and calculations[2] (typically by factors of two) are best attributed to differences in the incident neutron spectra.

The above exposure rates from $Na^{24}$ in the enclosure walls are negligible compared with the anticipated average exposure rate due to the synchrotron magnets themselves. In the Booster accelerator the exposure rate due to the magnets is 1.9 mrad/hr at a distance of 1 foot.[9] (The effects of deviations from average beam loss values and of injection losses, as discussed in Ref. 9, are not included here.) The smallness of the exposure rate from the walls is due in large measure to the low sodium content of the limestone concrete used.

REFERENCES

[1]P. J. Gollon, Radiation Levels in the Booster Enclosure, National Accelerator Laboratory Internal Report TM-97, Nov. 18, 1968.

[2]T. W. Armstrong and J. Barish, Calculation of. . .the Activation of Concrete by Neutrons. . ., Oak Ridge National Laboratory Report ORNL-TM-2630.

[3]1966 CERN-LRL-RHEL Shielding Experiment of the CERN Proton Synchrotron, Lawrence Radiation Laboratory Report UCRL-17941.

[4]P. J. Gollon, Radioactivation of the NAL Linac, National Accelerator Laboratory Internal Report TM-210, Feb. 12, 1970.

[5]H. Goldstein, The Attenuation of Gamma Rays and Neutrons in Reactor Shields, U. S. Government Printing Office, 1957.

[6]D. Nachtigall and S. Charalambus, Induced $Na^{24}$ Activity in the Concrete Shielding of High-Energy Accelerators, CERN ISR 66-28.

[7]M. Awschalom, T. Borak, and P. J. Gollon, Chemical Composition of Some Common Shielding Materials, National Accelerator Laboratory Internal Report TM-168, May 2, 1969, and results of later Na analyses.

[8]T. W. Armstrong and R. G. Alsmiller, Jr., Calculation of Residual Photon Dose Rate Around High-Energy Proton Accelerators, Oak Ridge National Laboratory Report ORNL-TM-2498, 1969.

[9]M. Awschalom, Lateral Shielding for the 8 GeV and 200 GeV Synchrotrons, National Accelerator Laboratory Internal Report TM-241, 1970.